

Incorporation of erbium ions into thin-film lithium niobate integrated photonics

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ABSTRACT

As an active material with favorable linear and nonlinear optical properties, thin-film lithium niobate has demonstrated its potential in integrated photonics. Integration with rare-earth ions, which are promising candidates for quantum memories and transducers, will enrich the system with new applications in quantum information processing. Here, we investigate the optical properties at $1.5 \mu\text{m}$ wavelengths of rare-earth ions (Er^{3+}) implanted in thin-film lithium niobate waveguides and micro-ring resonators. Optical quality factors near a million after post-annealing show that ion implantation damage can be repaired. The transition linewidth and fluorescence lifetime of erbium ions are characterized. The ion-cavity coupling is observed through a Purcell enhanced fluorescence from which a Purcell factor of 3.8 ± 0.5 , compared with waveguide lifetime, is extracted. This platform is compatible with top-down lithography processes and leads to a scalable path for controlling spin-photon interfaces in photonic circuits.

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As an important material in modern photonics, lithium niobate (LN) displays favorable piezoelectric, electro-optic, optical, photoelastic, and photorefractive properties.¹ It is widely used for electro-optic modulators, frequency doublers, optical parametric oscillators, and Q-switches for lasers. However, LN had been limited to bulk crystal components in all of these applications until a recent breakthrough in LN thin-film nanofabrication technology² made compact and dense photonic integrated circuits possible. Subsequent works on high-performance electro-optical modulators,^{3,4} ultra-efficient second harmonic generation,^{5,6} and microwave-optical transduction⁷ have stimulated intense interest and promise for integrated photonics. This advance in LN thin-film nanofabrication technology also raises the interest in incorporating rare-earth ions (REIs) into patterned LN waveguides and micro-cavities for scalable photonic integrated circuits with added functionalities enabled by the REIs.

REIs are well known for their applications in nonlinear optics such as lasers and amplifiers⁸ due to their stable optical transitions, high fluorescence quantum efficiencies, and long population lifetimes. Their narrow homogeneous linewidths,^{9,10} which allow the burning of ultra-narrow spectral holes, also find themselves useful in photonic signal processing¹¹ as well as frequency stabilization,^{8,12,13} medical imaging,¹⁴ and optical filtering.^{8,15} Over the past decades, REIs have

emerged as a promising candidate for quantum information processing,^{16–18} thanks to the weak coupling of their 4f electrons to the environment¹⁹ and long coherent spin states.²⁰ REIs have been used to demonstrate quantum memory protocols for quantum networks,²¹ light-matter interactions,²² and quantum-state teleportation.²³ Among the several well-studied REIs, erbium (Er) has received much attention due to its optical transitions in the telecommunications band, avoiding the need for frequency conversion and reducing the complexity of the system. There are already past efforts in incorporating erbium ions into various hosts for integrated photonics such as bulk LN,²⁴ yttrium orthosilicate (YSO) crystals,²⁵ and silicon nitride.²⁶ However, there is still a knowledge gap in combining erbium ions with LN thin-film structures, which could open a vital avenue to harness the benefit of both systems.

In this letter, we investigate the Er doping of LN photonic devices through ion implantation. We observe the recovery of the optical performance of the LN thin-film nanostructures post-annealing which repairs the lattice damage caused by implantation. The transition linewidth and fluorescence lifetime of the implanted ions inside optical waveguides are then characterized. The enhanced interaction of erbium ions with micro-cavities, which can be utilized to overcome the small oscillator strength of REIs, is demonstrated through the Purcell effect.

Commercial (NanoLN) thin-film wafers of 600 nm-thick z-cut lithium niobate on insulator (LNOI) are chosen for device fabrication and subsequent ion implantation. We fabricate micro-ring resonators and centimeter-long waveguides by patterning hydrogen silsesquioxane (HSQ) on LN thin films with electron beam lithography (EBL). Ring radius is designed to be 70 μm . The ring and the waveguide widths are 1.8 μm and 800 nm, respectively. The coupling gap between the ring and the bus waveguide is varied from 400 nm to 1000 nm to identify the critical coupling condition. Grating couplers optimized for the transverse electric (TE) mode at 1532 nm are used to efficiently screen many devices. The structures are then half-etched through reactive ion etching (RIE) with argon plasma. Figure 1(a)-(i) shows the scanning electron microscope (SEM) image of a grating coupler and Fig. 1(a)-(ii) shows the SEM image of the coupling region of a ring resonator. Figure 1(a)-(iii) shows the scanning electron microscope (SEM) image of a grating coupler patterned in LN. Figure 1(a)-(iv) shows the scanning electron microscope (SEM) image of the coupling region of a LN ring resonator.

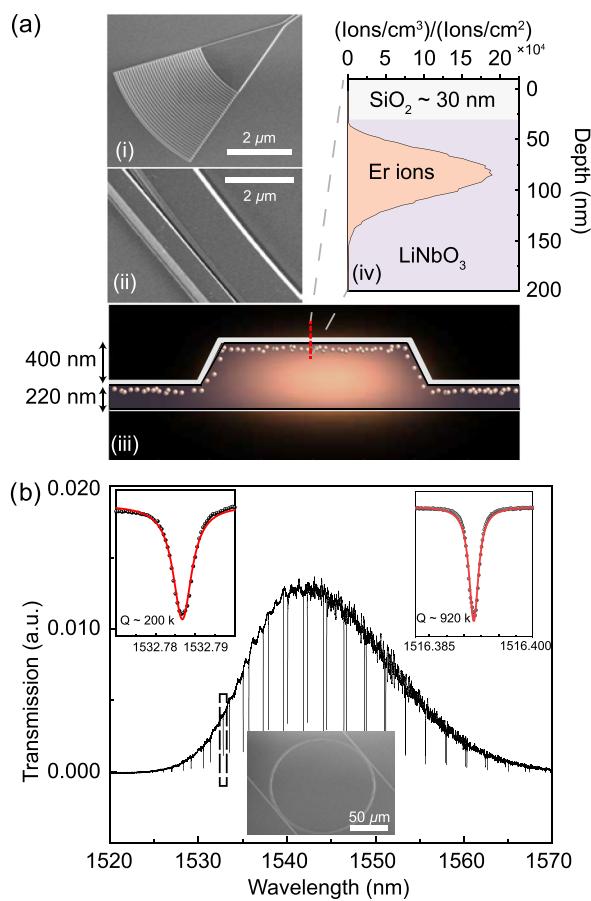


FIG. 1. (a) (i) SEM image of a grating coupler patterned in LN. (ii) SEM image of the coupling region of a LN ring resonator. (iii) Schematic cross section of a ring resonator and the electric field intensity distribution of the fundamental TE mode. The LN thin film is half-etched with a 60° vertical sidewall. (iv) SRIM simulation of the Er implantation depth distribution under a 350 kV acceleration voltage and a flux fluence of 1.14×10^{14} ions/cm². (b) The transmission measurement of a ring resonator. The two groups of modes correspond to the fundamental and first-order TE modes. The top-left inset is a resonance with a loaded Q of 200 k around 1532 nm indicated by the black-dashed box. The top-right inset is a resonance at 1516 nm in another resonator, showing a loaded Q of nearly a million. The bottom inset is a SEM image of a typical ring resonator with bus waveguides.

resonator. Figure 1(a)-(iii) shows a schematic of the cross section of a ring resonator with a 60° etched sidewall. The TE fundamental mode is well confined inside the half-etched ring. The ring resonators exhibit an average Q of 500 k and free spectral range (FSR) of 290 GHz after post-fabrication annealing at 500 °C for 5 h.

Before erbium implantation, 30 nm-thick SiO₂ is deposited on the chip by plasma-enhanced chemical vapor deposition (PECVD). The erbium ions are doped with an implantation energy of 350 keV and a flux fluence of 1.14×10^{14} ions/cm². The stopping and range of ions in matter (SRIM)²⁷ simulation [Fig. 1(a)-(iv)] estimates that the peak ion density is around 80 nm deep from the top surface with the tail of the distribution extending to the SiO₂/LiNbO₃ interface. This corresponds to an average implantation depth of 50 nm into the LN thin film.

Post-implantation annealing is carried out at various temperatures to repair the lattice damage from implantation. A temperature of 350 °C is sufficient to recover the transmission, but the maximum Q is not achieved until 550 °C. A typical grating coupler transmission measurement is shown in Fig. 1(b), in which fundamental and first-order TE modes of the resonator can be observed. The bottom inset of Fig. 1(b) shows a typical ring resonator with bus waveguides. A resonance with a Q of around 200 k at around 1532 nm in the device used for later experiments is shown in the inset at the top-left corner. It is worth noting that several high-Q resonances could be found in other ring resonators, one of which had a Q of nearly a million at 1516 nm, as shown in the inset at the top-right corner. The average Q of the devices after post-implantation annealing at 550 °C is 500 k, indicating that a full recovery of optical performance of the nanostructure is possible.

Figure 2(a) shows a cryogenic temperature (2.8 K) fluorescence measurement to map the inhomogeneous broadening of erbium ions. The experiment is carried out on a waveguide of 0.75 mm. A strong optical pump at 1480 nm is used to excite the Er³⁺ ions and the spectral response is mapped with a tunable filter. A reference laser is introduced to calibrate the tunable filter.

The crystal structure of LN is described by space group C_{3v}⁶, where Li⁺ and Nb³⁺ ions sit along the trigonal axis. When the Er³⁺ ions sit at the Li⁺ site, their point group is C₃.²⁸ This crystal field splits the Er³⁺ ground electronic state ($^4I_{15/2}$) into eight Kramer doublets labeled as Z₁₋₈ and the Er excited electronic state ($^4I_{13/2}$) into seven Kramer doublets labeled as Y₁₋₇. The optical pump at 1480 nm coincides with a strong absorption peak of the Z₁ – Y₆ transition.²⁹ Fast non-radiative decay³⁰ first relaxes the state to Y₁, and then optically relaxes to the ground Stark state Z₁ by emitting a photon at 1532 nm. This process is illustrated in the top-left inset of Fig. 2(a). The Er³⁺ fluorescence signal as well as the signature of the reference laser is shown clearly in Fig. 2(a). The Er³⁺ fluorescence peak is centered at around 1532 nm, and a Gaussian fit yields a linewidth of around 170 GHz. This linewidth is comparable to the literature value of 180 GHz for the low-temperature inhomogeneous broadening in bulk-doped crystals.⁸ The 10 GHz difference could be from the imperfect calibration of wavelengths based on the voltage input of the tunable filter. The device is then cooled down to 1.7 K, where a resonant excitation measurement is done to extract the population lifetime of the Er³⁺ ions. The inset of Fig. 2(b) shows the experiment schematic. Two acousto-optic modulators (AOM) are placed before and after the device for gating resonantly pump and measure the

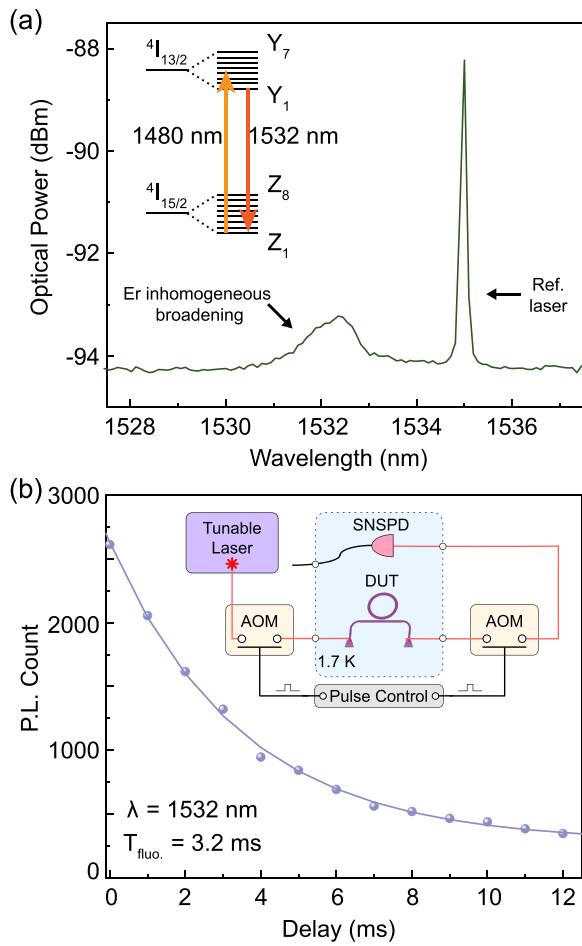


FIG. 2. (a) Non-resonant fluorescence measurement at 2.8 K with an optical pump at 1480 nm. A tunable filter is applied to collect the spectral response of the fluorescence around 1532 nm. A reference laser is combined via a wavelength division multiplexer to calibrate the tunable filter. The small peak near 1532 nm is due to the Er^{3+} fluorescence. The sharp peak at 1535 nm is due to the reference laser used for wavelength calibration. The relevant energy levels of Er:LN are shown in the top-left inset. The crystal field splits the $2S+1L_J$ states of the free Er^{3+} ions into to $J+1/2$ Kramer doublets that can be further split with an applied magnetic field. (b) Resonant fluorescence measurement at 1.7 K with an optical pump at 1532 nm. The fluorescence signal is gated and delayed for lifetime measurements. The solid line is a single exponential fit with a time constant of 3.2 ms. The inset shows the experimental schematic of the measurement.

fluorescence at 1532 nm using a continuous-wave tunable laser and a fiber-coupled superconducting nanowire single-photon detector (SNSPD).³¹ A single exponential fit of the fluorescence decay allows us to extract a lifetime of 3.2 ms. This is somewhat longer than the excited-state lifetime of 2.0 ms measured for a bulk-doped crystal by spectral hole burning.³² The exact cause of this lengthened lifetime is still uncertain and subject to further investigation.

To demonstrate the ion-cavity coupling, we focus on a resonance near 1532 nm in a ring resonator and extract the Purcell-enhanced fluorescence decay rate. The fluorescence decay when pumping on- and off resonance are compared in Fig. 3(a). Due to the interaction

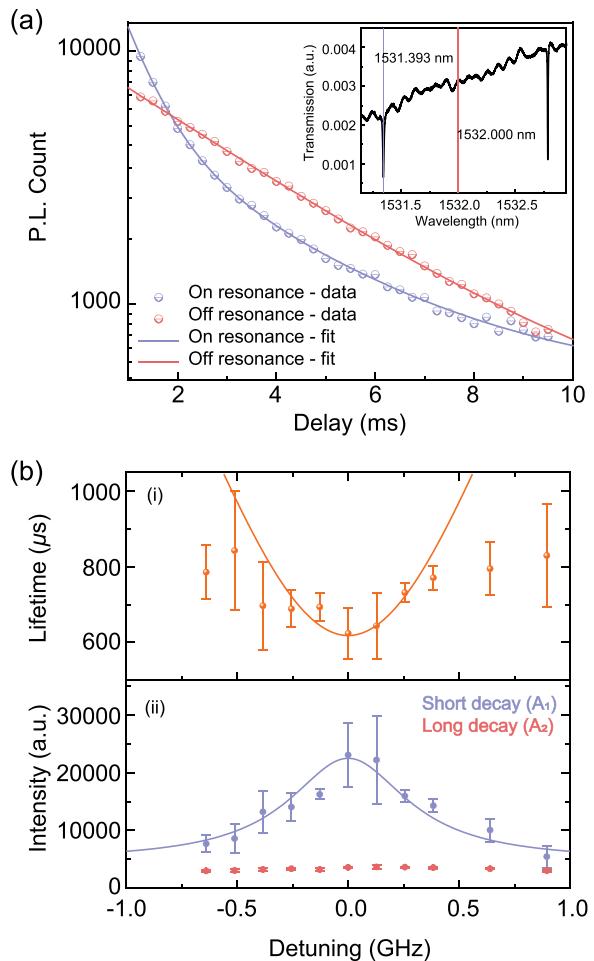


FIG. 3. (a) Fluorescence decay measurement on a semi-log scale for on-resonance pumping at 1531.393 nm (blue) and off-resonance pumping at 1532.000 nm (orange). The data from the off-resonance pump are fitted with a single exponential function, while those from the on-resonance pump are fitted with a double exponential function where the long decay time constant is fixed at the constant value obtained from the single-exponential fit of the fluorescence from a waveguide. The inset shows the transmission of the ring resonator near 1532 nm. The blue (orange) line indicates the on/off-resonance pump wavelength. (b) (i) Fluorescence lifetime from the short decay of the double exponential fit in (a). The solid line is the theoretical prediction based on the Purcell enhancement due to the ring resonance. (ii) The fluorescence intensities from the double exponential fit in (a). The short decay (blue) represents the fluorescence from the ring resonator and the long decay (red) represents the fluorescence from the bus waveguide. The solid line is the theoretical prediction based on the cavity enhancement.

with the cavity field, the fluorescence lifetime of an ion in the ring resonator (T_{ring}) should be shortened compared to the value in the bus waveguides outside of the cavity (T_{wg}), according to³³

$$\frac{1}{T_{\text{ring}}} = \frac{1}{T_{\text{wg}}} + \frac{\kappa g^2}{(\kappa/2)^2 + \delta^2}, \quad (1)$$

where $\kappa = \omega_0/Q$ is the decay rate of the ring resonator and $\delta = \omega - \omega_0$ is the detuning from the resonance frequency (ω_0). The coupling rate between the ion and resonator can be written as

$$g = \frac{\mu}{n} \sqrt{\frac{\omega}{2\epsilon_0 \hbar V}} \frac{|E(r)|}{|E_{\text{peak}}|}, \quad (2)$$

where μ , $n = 2.2$,³⁴ and $V = \frac{\int dV \epsilon(r) |E(r)|^2}{\max(\epsilon(r) |E(r)|^2)}$ are the dipole moment, refractive index, and resonator mode volume. The coupling also depends on the position of the ion, which is quantified by the spatial overlap with the cavity mode $|E(r)|/|E_{\text{peak}}|$. By substituting Eq. (2) into Eq. (1), we arrive at the formula for the Purcell factor at zero detuning

$$P = \frac{T_{\text{wg}}}{T_{\text{ring}}} - 1 = \frac{3}{4\pi^2} \left(\frac{\lambda_0}{n} \right)^3 \frac{Q}{V} \frac{|E(r)|^2}{|E_{\text{peak}}|^2}. \quad (3)$$

To quantify the coupling rate and the Purcell factor, we first fit the fluorescence measurement off resonance from the cavity with a single exponential decay function

$$f_{\text{off}} = A_0 \text{Exp} \left(-\frac{t}{T_{\text{wg}}} \right), \quad (4)$$

where $T_{\text{wg}} \approx 3$ ms is obtained. For the on-resonance case, however, there are two groups of ions contributing to the fluorescence, e.g., one in the ring resonator and the other in the bus waveguide. For simplicity, we approximate the influence of the cavity field with a Purcell enhanced lifetime averaged over all of the ions at different locations. Then the overall fluorescence from on-resonance pumping should be fitted with a double exponential decay function

$$f_{\text{on}} = A_1 \text{Exp} \left(-\frac{t}{T_{\text{ring}}} \right) + A_2 \text{Exp} \left(-\frac{t}{T_{\text{wg}}} \right). \quad (5)$$

Here, we fix the long time constant at $T_{\text{wg}} = 3$ ms since the waveguide fluorescence decay lifetime is constant over a wide range of wavelengths. As shown in Fig. 3(a), the experimental data are well approximated by this double exponential decay function, giving $T_{\text{ring}} = 618 \pm 68$ μ s. The average Purcell factor is then calculated to be $P_{\text{avg}} = 3.8 \pm 0.5$, and the coupling rate $g_{\text{avg}} = (2\pi) 597$ kHz.

The measured value of Purcell enhancement can be theoretically modeled by Eq. (3). For the selected resonance at 1531.393 nm, we measure $Q = 217$ k. To get the knowledge of field distribution inside the ring resonator, we conduct a mode analysis simulation in COMSOL software, whose result is shown in the color map of Fig. 1(a)–(iii). From the simulation, we calculate the mode volume of the fundamental TE mode $V = 223 \mu\text{m}^3$ and an average value of field overlap $|E(r)|^2/|E_{\text{peak}}|^2 = 0.125$ for the simulated implantation depth of 50 nm. This results in an overall Purcell enhancement $P = 3.1$, which is close to the experimental value of 3.8 ± 0.5 . We note that further improvement of the Purcell enhancement requires a resonance with a higher Q , a smaller mode volume V and a better spatial overlap $|E(r)|^2/|E_{\text{peak}}|^2$. For the current batch of devices, the enhancement could be about five times higher if the high- Q resonance was centered around the Er^{3+} transition wavelength of 1532 nm. Another factor of 8 improvement would also be possible if the Er^{3+} ions are incorporated into the center of the resonator so that full spatial overlap is achieved.

We also examine the fluorescence decay when the pumping frequency is detuned from the resonance. Figure 3(b)–(i) shows the measured lifetime of the ions in the ring resonator, extracted from the

double exponential fit, as well as the theoretical curve calculated based on Eq. (1). Apart from the experimental errors, the deviation from the theoretical curve could come from the uncertainty in the double exponential fitting. As the pump frequency is further detuned from the resonance, the fluorescence from the resonator becomes weaker, which undermines the accuracy of the fitting. Still, a trend of lifetime increase can be clearly observed. The corresponding fluorescence intensities from the ring and the bus waveguide portions are plotted in Fig. 3(b)–(ii). It is clear that the fluorescence from the waveguide (A_2) is insensitive to the detuning from the resonance, while the fluorescence from the ring resonator (A_1) peaks at zero detuning. Likewise, we theoretically model the enhanced fluorescence signal from the ions in the ring resonator (A_1) by a Lorentzian curve $A_1^{\text{theo}} = \frac{C}{(\kappa/2)^2 + (\delta)^2}$ with C being a fitting parameter. The model matches the data well in Fig. 3(b)–(ii), showing a clear resonance-enhanced fluorescence decay.

In conclusion, we introduce Er-implanted LN thin-film nanostructures and investigate their optical properties. We show that the cavity properties in the doped device could be recovered by post-implantation annealing with a temperature of up to 550 °C, obtaining resonances with a loaded Q of near a million after annealing. The transition linewidth and population lifetime of the erbium ions are extracted in waveguides and resonant cavities through fluorescence measurements. We show that the optical emission of ions in the ring resonator is cavity-enhanced and that the experimental data match theoretical models well, with a shortened lifetime of 618 μ s. This work demonstrates the possibility to fabricate a compact and scalable platform for spin-photon interfaces with LN-based integrated photonics.

Note added. In preparing this manuscript, the authors noticed a related pre-print discussing a different rare-earth element (Tm) in bulk-doped LN thin-film waveguides.³⁵

AUTHOR'S CONTRIBUTIONS

S.W. and L.Y. contributed equally to this work.

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